Activity measurements of the radionuclide ^{99m}Tc

for the NIST, USA in the ongoing comparison BIPM.RI(II)-K4.Tc-99m

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Abstract

In 2009, the first comparison of activity measurements of ^{99m}Tc using the Transfer Instrument of the International Reference System (SIRTI) took place at the National Institute of Standards and Technology (NIST), USA. An ampoule containing about 30 kBq of ^{99m}Tc traceable to the NIST primary standard was measured in the SIRTI for more than three half-lives. The influence of the ⁹⁹Mo impurity was negligible. The comparison, identifier BIPM.RI(II)-K4.Tc-99m, is linked to the BIPM.RI(II)-K1.Tc-99m comparison and the degrees of equivalence with the key comparison reference value and between the present NIST result and the 6 participants in the K1 comparison have been evaluated. Consistency between the USA and European activity measurements of ^{99m}Tc is demonstrated.

1. Introduction

Short-lived radionuclides are essential for most nuclear medicine where very short-lived (much less than one day) radionuclides are used. The use of nuclear medicine is increasing with the accessibility of these radionuclides which are consequently of great interest to the National Metrology Institutes (NMIs). However, sending ampoules of short-lived radioactive material to the Bureau International des Poids et Mesures for measurement in the International Reference System [1] is only practicable for the NMIs that are based in Europe. Consequently, to extend the utility of the SIR and enable other NMIs to participate, a transfer instrument has been developed at the BIPM with the support of the Consultative Committee for Ionizing Radiation CCRI(II) Transfer Instrument Working Group [2].

The BIPM ongoing K4 comparison of activity measurements of ^{99m}Tc (half-life $T_{1/2} = 6.0067$ h; u = 0.001 h [3]) has been set up in which the SIRTI, based on a well-type NaI(Tl) crystal calibrated against the SIR, is taken to the participating laboratory. The stability of the system is monitored using a ⁹⁴Nb reference source ($T_{1/2} = 20300$; u = 1600 a [4])¹ from the Institute for Reference Materials and Measurements (IRMM, Geel), which also contains the ⁹³Nb^m isotope. The ^{99m}Tc counting rate above a low-energy threshold, defined by the ^{93m}Nb x-ray peak at 16.6 keV, is measured relatively to the ⁹⁴Nb counting rate above the same threshold. Once the threshold is set, a brass liner is placed in the well to suppress the

¹ Hereafter, the last digits of the standard uncertainties are given in parenthesis.

^{93m}Nb contribution to the ⁹⁴Nb stability measurements. The ^{99m}Tc SIR ampoule is placed in the detector well with the brass liner to suppress the ^{99m}Tc x-ray peaks from the counts. No extrapolation to zero energy is carried out as all the measurements are made with the same threshold setting. The live-time technique using the MTR2 module from the Laboratoire National d'Essai – Laboratoire National Henri Becquerel, France (LNE-LNHB) [5] is used to correct for dead-time losses.

Similarly to the SIR, a SIRTI equivalent activity A_E is deduced from the ^{99m}Tc and ⁹⁴Nb counting results and the ^{99m}Tc activity measured by the NMI: A_E corresponds to the inverse of a calibration factor, i.e. the ^{99m}Tc counting rate in the TI divided by the activity of the source measured by the participant, where the ^{99m}Tc counting rate is expressed relatively to the ⁹⁴Nb counting rate. The possible presence of ⁹⁹Mo in the solution should be accounted for using γ -spectrometry measurements carried out by the NMI.

The protocol [6] for the BIPM.RI(II)-K4.Tc-99m comparison is available in the key comparison database of the CIPM Mutual Recognition Arrangement [7]. Publications concerning the details of the SIRTI and its calibration against the SIR are in preparation [8, 9].

2. Participant

As detailed in the protocol, participation in the BIPM.RI(II)-K4 comparisons is restricted to member states that are located geographically far from the BIPM and that have developed a primary measurement method for the radionuclide of concern. However, at the time of the comparison the National Metrology Institute (NMI) may decide for convenience to use a secondary method, for example a calibrated ionization chamber. In this case, the traceability of the calibration needs to be clearly identified.

After a successful trial comparison at the National Physical Laboratory (NPL), United Kingdom [9], the NIST became the first participant in the K4 comparison, which took place in May 2009. Through the calibration of the SIRTI against the SIR at the BIPM, this K4 comparison is linked to the BIPM.RI(II)-K1.Tc-99m comparison and thus degrees of equivalence between the NIST and all the K1 participants can be evaluated.

3. The SIRTI at the NIST

The reproducibility and stability of the SIRTI at the NIST have been checked by measuring the count rate produced by the reference 94 Nb source No. 1, the threshold position (defined by the 93m Nb x-ray peak), the background count rate, the frequency of the oscillator for the live-time correction and the room temperature as shown in Figure 1. The values indicated in the Figure are the differences from the values indicated in the figure description, using the appropriate units, as given, for each quantity measured.

The mean 94 Nb count rate, corrected for live-time, background and decay, measured at the NIST is 8493.5 (10) s⁻¹ which is in agreement with the mean since the set up of the system in

March 2007, 8492.7 (4) s⁻¹. In addition, the ⁹⁴Nb count rate has been checked on the return of the SIRTI to the BIPM after the comparison, giving the value of 8492.1 (16) s⁻¹. It should be noted that the uncertainty associated with the decay correction is negligible. The standard uncertainty associated with the live-time correction (effect of finite frequency of the oscillator) and the background is also negligible.



Figure 1: Fluctuation of the SIRTI at the NIST. Black squares: ⁹⁴Nb count rate above 8480 s⁻¹; circle: threshold position above 90 channels; stars: room temperature above 20 °C; open squares: background count rate above 48 s⁻¹; triangles: frequency of the oscillator above 10^6 Hz.

Although the threshold is tuned to correspond to the energy of the ^{93m}Nb x-ray peak, Figure 1 seems to show that the threshold position is anti-correlated with the ⁹⁴Nb count rate. The step in threshold position observed between the 19 May and 20 May corresponds to a slight change of the gain of the amplifier. However, the ^{99m}Tc measurements are much less sensitive to the threshold position [8]. Consequently, it was decided to use the mean ⁹⁴Nb count rate (instead of individual results) to normalize the ^{99m}Tc measurements so that the fluctuations observed in the ⁹⁴Nb results do not influence the ^{99m}Tc comparison results.

The room temperature was quite high (ca 25 °C) but relatively stable with an average temperature rise of 0.6 °C per day. It seems that the frequency of the oscillator for the live-time correction follows the temperature trend giving a frequency rise of 0.8 Hz per day. However such a small rise produces a negligible effect on the live-time correction and had no significant effect on the detector efficiency as the ⁹⁴Nb measurement results do not show any trend. Finally, it was verified by Monte-Carlo simulations using PENELOPE 2008 that the volume expansion of the source due to a 5 °C difference from the reference temperature of 20 °C has a negligible influence on the SIRTI efficiency.

4. The ^{99m}Tc solution standardized at the NIST

Details regarding the standardized solution are shown in Table 1, including any impurities, when present, as identified by the laboratory. The density and volume of the solution in the ampoule conform to the K4 protocol requirements².

Solvent / mol dm ⁻³	Carrier / μg g ⁻¹	Density at 20 °C / g cm ⁻³	Mass / g	⁹⁹ Mo impurity*
ШО	NoC1, 2040	1.004 (1)	3.598 9 (8)	$4.0(8) \times 10^{-6}$
H_2O	NaCl: 8940	Volume = $3.585 (4) \text{ cm}^3$		$4.0(8) \times 10$

Table 1: Characteristics of the ^{99m}Tc solution in ampoule D2

* Ratio of the ⁹⁹Mo activity to the ^{99m}Tc activity at the reference date

The ^{99m}Tc activity in the ampoule number D2 has been deduced from the measurement of the mother solution in an ionization chamber (IC) and a dilution factor of 265, resulting from two serial dilutions. The ionization chamber had been calibrated two months prior to the K4 comparison by live-timed $4\pi e X(PC)-\gamma(NaI)$ anti-coincidence counting (LTAC) with efficiency extrapolation [10]. The NIST measurement results are summarized in Tables 2 and 3 while the uncertainty budget of the IC calibration by primary measurement is given in appendix 2.

Table 2: ^{99m}Tc standardization at the NIST

Measurement method ACRONYM*	Activity / kBq	Standard uncertainty / kBq	Reference date YYYY-MM-DD	Half life used by the NIST / h
IC calibrated in March 2009 by ^{4P-IC-GR-00-00-00} 4πeX(PC)-γ(NaI) anticoincidence ^{4P-PP-MX-NA-GR-AC}	31.81	0.14	2009-05-21 17:00 UTC	6.006 7 (10)

* See appendix 1

 $^{^{2}}$ At the time of the comparison, the protocol required 3.6 (2) g of solution in the ampoule. The NIST ampoule conforms to the new protocol requirement of 3.6 (1) cm³.

Uncertainty contributions due to	Comments	Evaluation method	Relative standard uncertainties × 10 ⁴
Ionization chamber measurement variability	Standard deviation of the mean for 3 values. Each value represents 1 ampoule inserted 3 times into Chamber A with 10 charge measurements per insertion, relative to the radium reference source, inserted 4 times.	A	0.8
Use of ionization chamber	Estimated uncertainty due to source positioning and variability of ampoule thickness and solution density between IC calibration and present measurements.	В	5
Gravimetric links	Estimate of uncertainty in dilution factor to solution D1 and mass of active solution contained in ampoule D2, based on agreement between dispensed and contained masses and previous studies of mass measurement accuracy.	В	4.7
^{99m} Tc half-life	Uncertainty due to decay correction from time of IC measurements to reference time.	В	0.6
Ionization chamber calibration (see appendix 2)	Combined uncertainty for IC A calibration factor including primary standardization by LTAC method and linkage to IC A.	В	42
⁹⁹ Mo impurity	Uncertainty of impurity correction to IC activity determination, based on $^{99}Mo/^{99m}Tc$ impurity ratio of 4.0 (0.8) × 10 ⁻⁶ on the reference date, as determined by HPGe γ -ray spectrometry. No other impurities found.	В	0.02
R		43	

Table 3: NIST uncertainty budget for the activity measurement of ampoule D2 (May 2009)

5. The ^{99m}Tc measurements in the SIRTI at the NIST

The maximum count rate corrected for live-time in the NaI(Tl) was 23 150 s⁻¹ which is over the limit of 20 000 s⁻¹ set in the protocol [6]. Consequently, the first four measurements were discarded.

In principle, the live-time correction should be modified in order to take into account the decaying count rate [11]. In the present experiment, the measurement duration has been limited to 1800 s so that the relative effect of decay on the live-time correction is less than 10^{-4} .

After the first series of 99m Tc measurements, the presence of 11 droplets of ca 1.5 mm diameter was noted on the inner walls of the ampoule, in the cylindrical part and in the ampoule head. As shaking the ampoule manually did not remove these droplets, it was decided to centrifuge the 99m Tc ampoule for 5 min. at 3000 r/min. The centrifuging suppressed the droplets on the walls and great care was taken to keep the ampoule always vertical afterwards. The measurement results defined in the protocol by a SIRTI equivalent activity A_E

differ by 1.7×10^{-3} after centrifuging as shown in Figure 2. Such an influence of droplets on the ampoule walls was confirmed by Monte-Carlo simulation using PENELOPE [8] and consequently the first series of measurements was discarded from the final analysis.



Figure 2: The ^{99m}Tc measurement results in the SIRTI. A decrease in SIRTI equivalent activity A_E (i.e. an increase of detection efficiency) is observed after centrifuging at t = 4 h. The NIST ^{99m}Tc measurement uncertainty and the ⁹⁴Nb mean count rate uncertainty, which are both constant over all the measurements, are not included in the uncertainty bars shown on the graph.

The correction for the ⁹⁹Mo impurity mentioned in Table 1 is negligible even after one day of measurements when the ^{99m}Tc had decayed by a factor of 15. This is a consequence of having diluted the mother solution quickly after its production in order to obtain an ampoule measurable in the SIRTI as soon as possible, instead of waiting for the ^{99m}Tc decay with a growing influence of the longer-lived ⁹⁹Mo.

The time of each SIRTI measurement was obtained from the synchronization of the laptop with a time server in the USA. Unfortunately, the connection with the server was interrupted during the measurements, producing an error of 1 s in the measuring time. However, this error has a negligible effect on the results.

The uncertainty budget for the SIRTI measurements of the ^{99m}Tc ampoule is given in Table 4. More detail can be found in reference [8].

Uncertainty contributions due to	Comments	Evaluation method	$\begin{array}{c} \textbf{Relative standard} \\ \textbf{uncertainties} \\ \times 10^4 \end{array}$
⁹⁴ Nb measurement including threshold setting	Weighted standard deviation of 6 series, each series consisting of 10 measurements	А	1.1
Long-term stability of the SIRTI	Weighted standard deviation of 25 series, each series consisting of 10 measurements	А	0.4
^{99m} Tc measurement including live-time, background and decay corrections	Standard uncertainty of the weighted mean of 56 measurements, taking into account the correlation due to the ^{99m} Tc half-life.	А	2.6
Ampoule dimensions	From IRMM report [12] and sensitivity coefficients from Monte-Carlo simulations	В	7
Ampoule filling height	Solution volume requested in the protocol is 3.6 (1) cm ³ ; sensitivity coefficients from Monte-Carlo simulations	В	6
Solution density	Between 1 g/cm ³ and 1.01 g/cm ³ as requested in the protocol; sensitivity coefficients from Monte-Carlo simulations	В	0.8
Rela	9.7		

6. Comparison result and degrees of equivalence

The comparison result is taken as the weighted mean of all the $A_{\rm E}$ values after centrifuging the ampoule. The standard uncertainty $u(A_{\rm E})$ is obtained by adding quadratically the SIRTI combined uncertainty from Table 4 and the uncertainty quoted by the NIST for the ^{99m}Tc measurement (see Table 2). The correlation between the NIST and the BIPM due to the use of the same ^{99m}Tc half-life is negligible in view of the small contribution of this half-life to the combined uncertainty of the NIST ionization chamber measurement. The K4 comparison result is given in Table 5 as well as the linked result $A_{\rm e}$ in the BIPM.RI(II)-K1.Tc-99m comparison which was obtained by multiplying $A_{\rm E}$ by the linking factor L = 12 174 (19). The linking factor was obtained through the measurement of three ^{99m}Tc ampoules from the LNE-LNHB and the NPL in both the SIRTI and the SIR [9].

Reference date YYYY-MM-DD	Measurement method ACRONYM*	Solution volume /cm ³	A _E /kBq	u(A _E) /kBq	Linked A _e /kBq	u(A _e) /kBq
2009-05-21 17:00 UTC	Ionization chamber ^{4P-IC-GR-00-00-00} calibrated by 4πeX(PC)-γ(NaI) anticoincidence ^{4P-PP-MX-NA-GR-AC}	3.584 (4)	12.554	0.057	152 840	730

Table 5: BIPM.RI(II)-K4.Tc-99m comparison result and link to the BIPM.RI(II)-K1.Tc-99m comparison

* See appendix 1

Every participant in the K4 comparison is entitled to have one result included in the key comparison database (KCDB) as long as the laboratory is a signatory or designated institute listed in the CIPM MRA. Normally, the most recent result is the one included. Any participant may withdraw its result only if all the participants agree.

The key comparison reference value (KCRV) for ^{99m}Tc has been defined in the frame of the BIPM.RI(II)-K1.Tc-99m comparison using direct contributions to the SIR. The most recent updated value is 153 240 (220) kBq as detailed in reference [13].

The degree of equivalence of a particular NMI, i, with the KCRV is expressed as the difference D_i with respect to the KCRV

$$D_i = A_{e_i} - \text{KCRV} \tag{1}$$

and the expanded uncertainty (k = 2) of this difference, U_i , known as the equivalence uncertainty, hence

$$U_i = 2u_{D_i}, \qquad (2)$$

taking correlations into account as appropriate [14].

The NIST K4 result agrees with the KCRV within a standard uncertainty.

The degree of equivalence, D_{ij} , between any pair of NMIs, *i* and *j*, is expressed as the difference in their results

$$D_{ij} = D_i - D_j = A_{e_i} - A_{e_j}$$
(3)

and the expanded uncertainty of this difference U_{ij} where

$$u_{D_{ij}}^{2} = u_{i}^{2} + u_{j}^{2} - 2u(A_{e,i}, A_{e,j})$$
(4)

where any obvious correlations between the NMIs (such as a traceable calibration) are subtracted using the covariance $u(A_{ei}, A_{ej})$, as are normally those correlations coming from the SIR and the SIRTI.

The uncertainties of the differences between the values assigned by individual NMIs and the KCRV are not necessarily the same uncertainties that enter into the calculation of the uncertainties in the degrees of equivalence between a pair of participants. Consequently, the uncertainties in the table of degrees of equivalence cannot be generated from the column in the table that gives the uncertainty of each participant with respect to the KCRV. However,

the effects of correlations have been treated in a simplified way as the degree of confidence in the uncertainties themselves does not warrant a more rigorous approach.

Table 6 shows the matrix of all the degrees of equivalence as they appear the KCDB. It should be noted that for consistency within the KCDB, a simplified level of nomenclature is used with A_{ei} replaced by x_i . The introductory text is that agreed for the comparison. The graph of the first column of results in Table 6, corresponding to the degrees of equivalence with respect to the KCRV (identified as x_R in the KCDB), is shown in Figure 3. The graphical representation indicates in part the degree of equivalence between the NMIs but does not take into account the correlations between the different NMIs. However, the matrix of degrees of equivalence shown in yellow in Table 6 does take the known correlations into account.

Conclusion

The BIPM ongoing key comparison for ^{99m}Tc, BIPM.RI(II)-K4.Tc-99m currently comprises one result (NIST) that has been linked to the BIPM.RI(II)-K1.Tc-99m comparison. This first K4 result has been analysed with respect to the KCRV determined for this radionuclide in the frame of the K1 comparison, and with respect to the other six results of the K1 comparison. The matrix of degrees of equivalence has been approved by the CCRI(II) and is published in the BIPM key comparison database.

The agreement of this first K4 result with the KCRV supports the successful implementation of the SIR transfer instrument and its robustness following overseas transportation.

Other results may be added as and when other NMIs contribute ^{99m}Tc activity measurements to the K4 or K1 comparisons or take part in other linked Regional Metrology Organization comparisons.

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 Table 6.
 Table of degrees of equivalence and introductory text for ^{99m}Tc

Key comparison BIPM.RI(II)-K1.Tc-99m

MEASURAND : Equivalent activity of ^{99m}Tc

Key comparison reference value: the SIR reference value for this radionuclide is x_R = 153.2 MBq, with a standard uncertainty u_R = 0.2 MBq.

 x_{R} is computed as the mean of the results obtained by primary methods.

The degree of equivalence of each laboratory with respect to the reference value is given by a pair of terms: $D_i = (x_i - x_R)$ and U_i , its expanded uncertainty (k = 2), both expressed in MBq, with n the number of laboratories, $U_i = 2((1-2/n)u_i^2 + (1/n^2)\Sigma u_i^2)^{1/2}$ when each laboratory has contributed to the reference value (see Final Report).

Linking BIPM.RI(II)-K4.Tc-99m to BIPM.RI(II)-K1.Tc-99m

The value x_i is the SIRTI equivalent activity for laboratory *i* participant in BIPM.RI(II)-K4.Tc-99m multiplied by the linking factor to BIPM.RI(II)-K1.Tc-99m (see Final report).

The degree of equivalence of laboratory *i* participant in BIPM.RI(II)-K4.Tc-99m with respect to the key comparison reference value is given by a pair of terms: $D_i = (x_i - x_R)$ and U_i , its expanded uncertainty (k = 2), both expressed in MBq. The approximation $U_i = 2(u_i^2 + u_R^2)^{1/2}$ is used in the following table.

The degree of equivalence between two laboratories *i* and *j*, one participant in BIPM.RI(II)-K1.Tc-99m and one in BIPM.RI(II)-K4.Tc-99m, or both participant in BIPM.RI(II)-K4.Tc-99m, is given by a pair of terms: $D_{ij} = D_i \cdot D_j$ and U_{ij} , its expanded uncertainty (k = 2), both expressed in MBq, where the approximation $U_{ij} = 2(u_i^2 + u_j^2 - 2x_ix_j(u_i/l)^2)^{1/2}$ is used with u_i being the uncertainty of the linking factor *I* when both laboratories are linked.

These statements make it possible to extend the BIPM.RI(II)-K1.Tc-99m matrices of equivalence to the other participants in BIPM.RI(II)-K4.Tc-99m



Figure 3. Graph of degrees of equivalence with the KCRV for ^{99m}Tc



N.B. The right-hand axis gives approximate relative values only

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Appendix 1 Acronyms used to identify different measurement methods

Each acronym has six components, geometry-detector (1)-radiation (1)-detector (2)-radiation (2)-mode. When a component is unknown, ?? is used and when it is not applicable 00 is used.

Geometry	acronym	Detector	acronym
4π	4P	proportional counter	PC
defined solid angle	SA	press. prop. counter	PP
2π	2P	liquid scintillation counting	LS
undefined solid angle	ndefined solid angle UA Nal(TI)		NA
		Ge(HP)	GH
		Ge(Li)	GL
		Si(Li)	SL
		CsI(TI)	CS
		ionization chamber	IC
		grid ionization chamber	GC
		bolometer	BO
		calorimeter	CA
		PIPS detector	PS
Radiation	acronym	Mode	acronym
positron	PO	efficiency tracing	ET
beta particle	BP	internal gas counting	IG
Auger electron	AE	CIEMAT/NIST	CN
conversion electron	CE	sum counting	SC
mixed electrons	ME	coincidence	со
bremsstrahlung	BS	anti-coincidence	AC
gamma rays	GR	coincidence counting with efficiency tracing	СТ
X - rays	XR	anti-coincidence counting with efficiency tracing	AT
photons (x + γ)	PH	triple-to-double coincidence ratio counting	TD
photons + electrons	PE	selective sampling	SS
alpha - particle	AP	high efficiency	HE
mixture of various radiations	MX	digital coincidence counting	DC

Examples method	acronym
$4\pi(PC)\beta$ - γ -coincidence counting	4P-PC-BP-NA-GR-CO
$4\pi(PPC)\beta-\gamma$ -coincidence counting eff. trac.	4P-PP-MX-NA-GR-CT
defined solid angle α -particle counting with a PIPS detector	SA-PS-AP-00-00-00
4π (PPC)AX- γ (Ge(HP))-anticoincidence counting	4P-PP-MX-GH-GR-AC
4π CsI- β ,AX, γ counting	4P-CS-MX-00-00-HE
calibrated IC	4P-IC-GR-00-00-00
internal gas counting	4P-PC-BP-00-00-IG

Uncertainty contributions due to	Comments	Evaluation method	Relative standard uncertainties × 10 ⁴
Ionization chamber measurement variability	Standard deviation of the mean for 18 values: 6 ampoules inserted 3 times each, measured relative to the radium reference source inserted 8 times.	A	0.6
Ionization chamber density correction	Uncertainty in measured ratio (1.0032) of chamber A response for saline ($\rho = 1.004$ g/mL) compared to 3 M HCl ($\rho = 1.048$ g/mL).	В	7
Gravimetric links	Estimate of uncertainty in dilution factors and masses of point sources beyond variability contained in other listed components.	В	2
^{99m} Tc half-life	Uncertainty due to half-life for 99m Tc in dried point sources. Using (6.014 h; $u = 0.006$ h), which is average of pertechnetate (6.006 7 h) and sulfide (6.021 h). VYNS [#] films were dried in H ₂ S atmosphere, but unsure how much sulfide was formed.	В	9.5
⁹⁹ Mo impurity	Uncertainty of impurity correction of $< 10^{-4}$ to ionization chamber activity measurements, as determined by HPGe γ -ray spectrometry. No other impurities found.	В	0.3
LTAC measurement variability	Weighted standard deviation of the mean for 13 determinations encompassing 4 sources and 2 background measurements. All determinations used nominally the same efficiency extrapolation domain.	А	16
Efficiency extrapolation	Typical standard deviation of the distribution for extrapolations using 3 different, reasonable, efficiency domains.	В	35
Live-time	Experimental limit on live-time accuracy based on previous measurements of sources of various strengths, measurements with various extending dead-times and tests of the oscillator accuracy.	В	10
R [#] Copolymer of chlori	elative combined standard uncertainty		42

Appendix 2: Uncertainty budget for the ionization chamber calibration by anticoincidence primary measurements (March 2009)

[#] Copolymer of chloride and vinyl acetate